Prompt fission neutron spectra and angular distributions measured in narrow windows of fragment masses and total kinetic energies

Puzzling results and an eventual explanation

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Plan

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Motivation

Dynamical calculations of the fission process correspond to a given fission path at a time, leading to a given scission configuration. To compare with inclusive experimental data one has to integrate over all possible scission configurations which is rarely possible. The best one can do is to restrict the number of configurations, e.g., by selecting data for a given fragment mass ratio and TKE, a condition that requires the collection of a large total number of fission events.

Moreover, non-inclusive data are expected to reveal new features that are washed out when averaging over many scission paths.

Such data are needed to elucidate current fission topics and could be obtained by measurements in coincidence with perfectly separated (in Z and A) fission fragments (provided by SOFIA or VAMOS) having in addition well defined total kinetic energies (TKE). An example would be measurements of PFN angular distributions with respect to the fission axis, PFN kinetic energy spectra and PFN multiplicities

Precise existing data

The ideal experiment mentioned above has no chances to be performed soon since it requires an extremely large statistics and a very complex experimental setup.

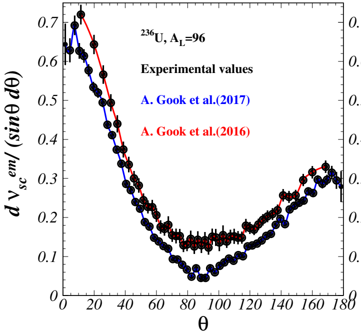
Until then, in the present study, suitable PFN data for 236U and 252Cf, obtained recently by Gook at al. at JRC-Geel, are used.
Prompt fission neutrons (PFN) angular and energy distributions were measured in coincidence with fission fragments. Improved angular, mass and energy resolutions as well as very good statistics have been achieved.

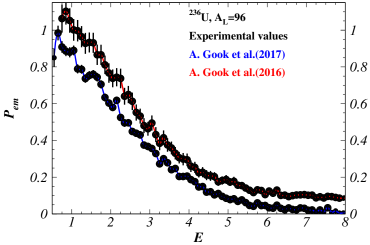
In a first campaign [1], the reaction ^{235}U(n,f) was studied. The angular, fragment-mass (pre-neutron) and TKE resolutions (FWHM) were 9.5 deg, 4.9 amu and 0.9 MeV respectively.

In a second campaign, a previous study of ^{252}Cf(sf) [3] was repeated using an improved setup.

Ang and en distr for a given mass ratio (A_L=96); ^{235}U(n,f)

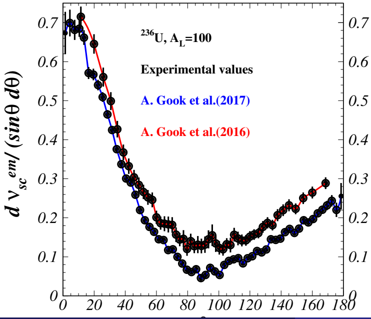
When windows on fragment masses were put ($A_L \pm 2$), the measured distributions exhibit fine structures. A_L=96 defines the most probable mass division for ^{236}U. The TKE values were not restricted.

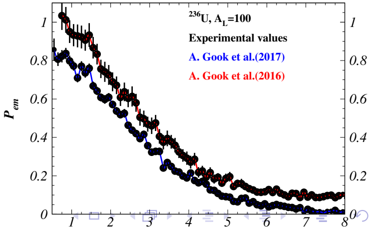




This result was puzzling but it was at the limit of the statistical errors. The experiment was therefore repeated in a second campaign [2] with better statistics (10^6 coincident events) and a different setup to exclude that scattering of neutrons on objects inside the reaction chamber is the cause for these structures. The detectors' position was changed. As a result, see lower curves (2017), the structures were reproduced and became more clear. It seems they are real. The red and blue curves are shifted to allow the comparison of their fine structures.

The results are similar for all mass ratios. See below for A_L=100.





A harsh judgement

At first sight, these identified structures in the data are not compatible with the traditional hypothesis that PFN are evaporated from fully accelerated fragments. This statement made many eyebrows raise. It is always more comfortable to follow the conventional wisdom. How could we be wrong for so long time (80 years)?

To resolve the doubts about the origin of PFN emission, that these new data raised, there are two directions to follow. One is to repeat the experiment and make sure that indeed the distributions are not smooth. The second is to find an alternative mechanism for the emission of prompt neutrons during fission which leads to non-smooth distributions.

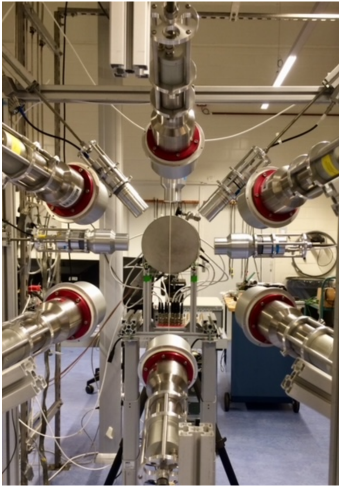
In this talk we are presenting our contributions in each of these directions.

Make a better experiment

Are the structures outside the statistical errors? To answer this unavoidable question, a previous study of ^{252}Cf(sf) [3] was repeated using an improved setup. Since a neutron beam is not needed, one can collect a larger number of events than in ^{235}U(n,f). In ref.[3], there was only one liquid scintillator neutron detector placed along the normal to the target. To limit the energy loss in the target, fission events with large angles between the fission axis and the neutron detector were rejected leading to a limitation of the effective solid angle.

In the new experiment six neutron detectors, with an azimuthal angular distance of 60 degree, placed around the chamber allowing measurements with good mass resolution (4.2 amu) at any angle with respect to the fission axis. A position-sensitive Frisch-grid ionization chamber was used to detect the fission fragments. The experiment lasted 3 months. 68×10^6 coincident events were collected.

^{252}Cf experimental setup



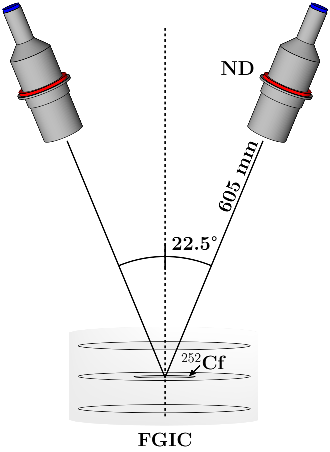
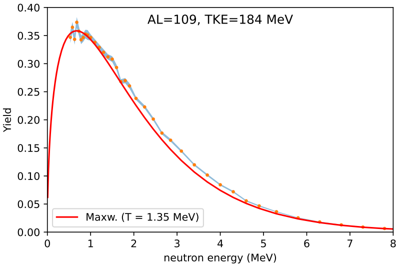
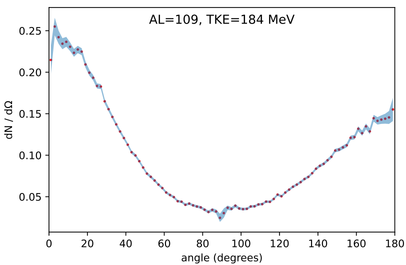


Figure: Illustration of the experimental setup. On the left-hand side, a photo taken from behind the neutron detector arrays is shown. The right-hand side shows a cross-section through the setup, where relevant dimensions are given.

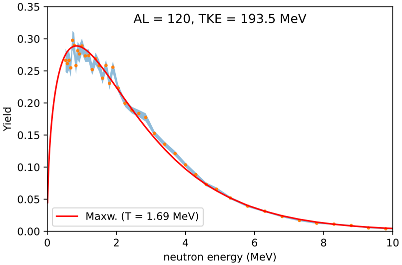
New ^{252}Cf data: PFN spectrum and angular distribution for A_L=109±2 and TKE=184.0±1 MeV

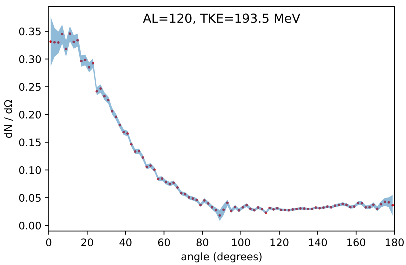




The chosen configuration represents the most probable fragment mass A_L and the corresponding TKE. The statistics is the highest among other fragmentations. The uncertainty is marked by the blue ribbon. At 0 and 180 it increases a bit, but for the rest it is the same as the point size. Not much room for doubts is left: in both distributions there are deviations from the smooth curves expected if neutrons are evaporated.

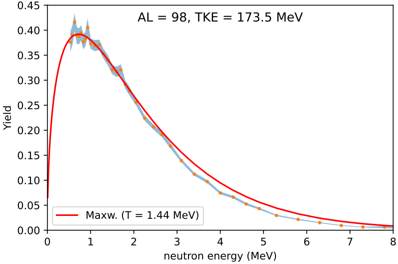
New ^{252}Cf data: PFN spectrum and angular distribution for A_L=120 and TKE=193.5 MeV

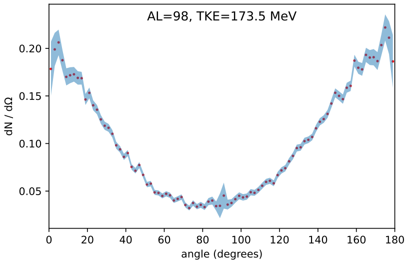




This fragmentation (120,132) corresponds to the double magic A_H=132. It has also a high yield. It is worth noticing the similarities between the structures in this case and the previous one, i.e., $A_L = 109$ and 120. In both cases, deviations from a Maxwellian spectrum were found from 0.5 to 6 MeV. They consist in fine structures (wiggles), more pronounced around the most probable energy ($\approx$1 MeV).

New ^{252}Cf data: PFN spectrum and angular distribution for A_L=98 and TKE=173 MeV





This 3rd fragmentation (98,154) is the most asymmetric. The neutrons emitted in the direction of the heavy fragment have a higher yield. It is worth noticing the similarities between the structures in all three cases, i.e., $A_L = 98$, 109 and 120.

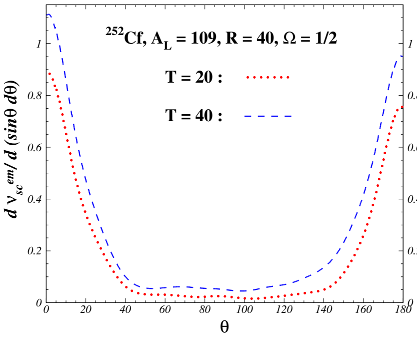
Similar deviations from a Maxwellian spectrum were found. They consist in fine structures (wiggles) around the most probable energy (≈1 MeV).

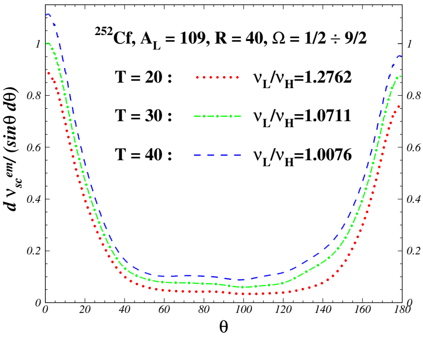
Alternative mechanism: dynamical emission during scission

An alternative to the statistical model is the dynamical scission model (DSM) [4]. The emission of scission neutrons (SN) is due to the diabatic coupling between the neutron degree of freedom and the changing neutron-nucleus potential during the scission process (i.e., from the neck rupture at finite radius r_{min} to the absorption of the neck stubs by the fragments). This tiny diabatic part of the fission process was investigated using the time-dependent Schrödinger equation with time-dependent potential.

DSM was applied recently to the calculation of the PFN angular distribution with respect to the fission axis [5] and of the PFN energy spectrum [6]. The calculation was performed for the most probable scission configuration. ^{236}U simulating the reaction ^{235}U(n$_{th}$,f) was taken as example. It was shown that, if neutrons are released during the neck rupture, the distributions present oscillations (structures) due to both the proximity of the fragments at the instant of emission and the finite number of neutrons that contribute.

Typical angular distributions predicted by DSM





The angular distributions display weak oscillations (from 50° to 150°). They could be the sign of scattering of neutrons on the just born fragments [5]. The maxima and minima are typical of a non-monotonic deflection function (rainbow effect).

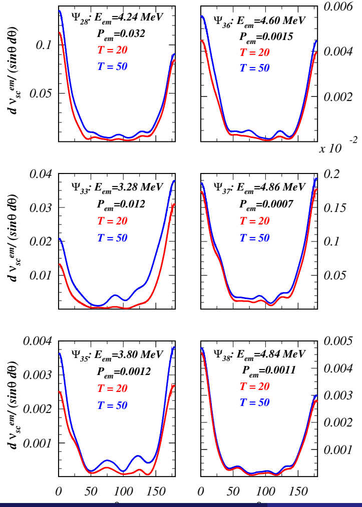
Angular distribution - formula

The angular distribution is given by the integral with respect to time of the number of neutrons that leave a sphere of radius R (around the fissioning nucleus) in a solid angle $d\Omega$ and in a time interval dt
$d\nu_{sc}^{em} = \bar{J}_{em}(R,\theta,t)\bar{n}(R,\theta,t)R^2 dt d\Omega.$
The current density $\bar{J}_{em}(\rho,z) = \frac{i\hbar}{\mu}\sum_i v_i^2(f^i\bar{\nabla}f^{i*} - f^{i*}\bar{\nabla}f^i)$ with $f^i = |\Psi_{em}^i\rangle$, provides the distribution of the average directions of motion of the unbound neutrons at t=T. The upper limit should in principle be ∞.

Due to the diabaticity of the scission process, each neutron of the fissioning nucleus is more or less emitted and therefore each contributes to the angular (and energy) distributions. The angular distributions are very different from one state to another but all strongly oscillate.

When summing over all states the oscillations are reduced. This is the reason why the amplitudes of the oscillations in the total distribution are small.

Angular distributions for single neutron states. They are different from one state to another and all strongly oscillate



Angular distributions for single neutron states with Ω=1/2 at 2 times T= 20 and 50 $\times$ 10^{-22}sec. P_{em} is the emission probability of each state.

Most of them are peaked in the direction of the L-fragment but some prefer the H-fragment and few move with equal probability in both directions

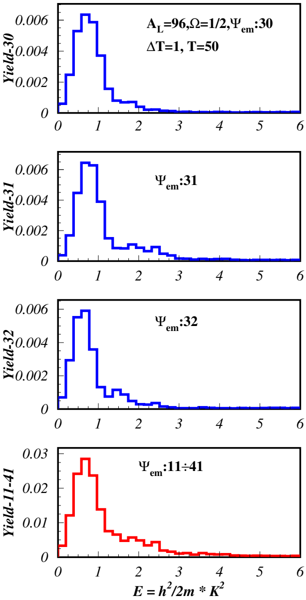
Energy spectrum of the scission neutrons

It is the most difficult to calculate since it is necessary to propagate in time the unbound part $|\Psi^i_{em}\rangle$ of each neutron wave packet until it completely leaves the fissioning system. It is a hard numerical task that requires very large (ρ,z) grids and very long CPU times. We were able to go until $\Delta T + T_{max}$ with $T_{max} = 50 \times 10^{-22}$ sec.
The Fourier transforms of these wave packets

$$\mathrm{F}^i(k_\rho, k_z, T) = 2\pi \int_{-\infty}^{\infty} \left[\int_0^\infty \Psi^i_{em}(\rho, z, T) J_0(2\pi\rho k_\rho)\rho \mathrm{d}\rho\right] \mathrm{e}^{-2\pi \mathrm{i} z k_z} \mathrm{d}z$$

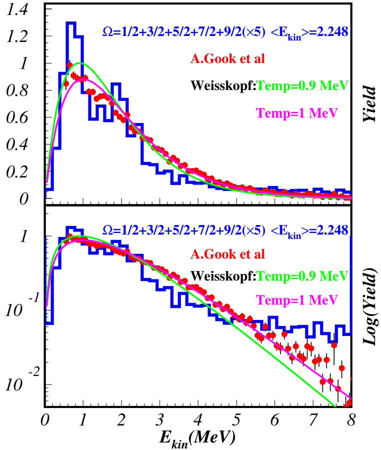
are calculated in order to get the corresponding momentum distributions which lead to the kinetic energy distributions. To obtain the whole kinetic energy spectrum for a fixed mass asymmetry, one has to sum the single spectra over all occupied states and all Ω values.

Kinetic energy distribution for Ω=1/2 neutron states



The total distribution (red/bottom) is a finite weighted sum (30) of individual quasi-maxwellian distributions with different mean values and widths. Three such examples are plotted (blue/above).
For this reason the spectrum cannot be smooth.

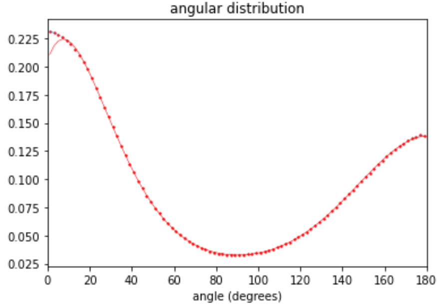
Total (all Ω values) Scission-Neutron Spectrum for ^{236}U

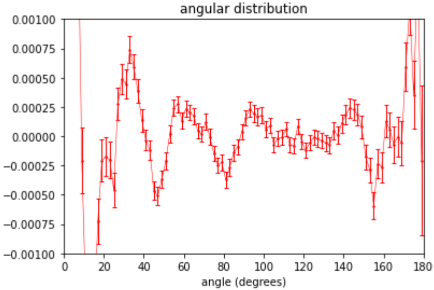


The calculated kinetic energy spectrum for the most probable mass division (defined by the light fragment mass AL = 96) (histogram) is compared with experimental data from the reaction ^{235}U(n,f). Two typical evaporation spectra characterized by nuclear temperatures 1.0 and 0.9 MeV are also shown.

A closer look at the high energy tail of the spectrum shows the data lying between scission and evaporated neutrons. One could speculate that the scission neutrons amount to approximately half of PFN.

^{252}Cf angular distribution without any selection in fragment mass or TKE: another puzzling result.





Left: polynomial fit to data. The chosen order of the polynom (7) lead to the minimum chi square per degree of freedom.

Right: the residual of the polynomial fit shows oscillations (in the region from 20° to 160°) in spite of using a relatively high order polynomial. They are at 1% level but they are statistically significant.

⇒ There are always oscillations, even in the inclusive PFN data.

Conclusions

There are structures both in the data and in the calculations. More work is necessary in order to asses if they have or not the same origin.

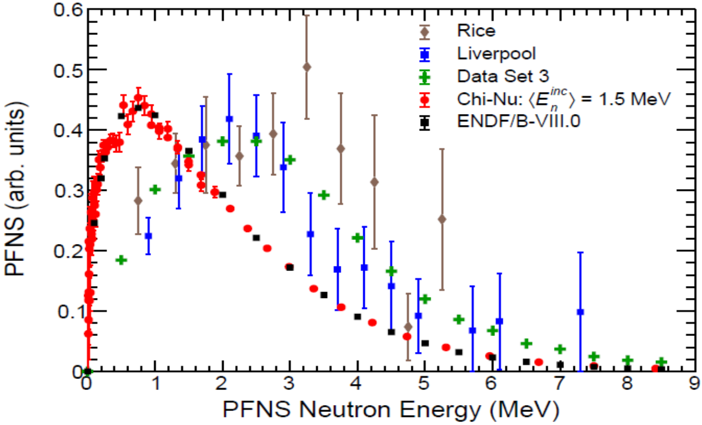
From the experimental side:
1) We could try to obtain a better mass resolution but we are already close to the limit imposed by neutron emission. The two masses are deduced from their measured energies imposing energy and momentum conservation. Being post neutron energies they have to be corrected for the recoil.
2) The angular resolution (9-10 degree FWHM) is limited by the opening angle of the detectors. We could get it down to that of the chamber (7 degree FWHM) by moving the detectors further away, from 60 cm to 120 cm.
3) For the spectrum, we could lower the neutron detection threshold using other type of detectors.

Conclusions (cont.)

4) Repeating the experiment for thermal neutron induced fission of ^{235}U or ^{239}Pu has its advantages. Large statistical accuracy could be obtained, because of the large thermal neutron cross-section, at facilities that can provide large thermal flux (ILL-Grenoble). The lower average number of neutrons emitted in ^{236}U would also benefit the mass resolution which can be obtained with the 2E technique, since the recoil correction is smaller.

From the theoretical side:
1) More results are needed especially for ^{252}Cf.
2) DSM contains approximations; they should be tested.
3) An alternative program should be developed with new numerical algorithms and a new shape parametrization.

Other recent data showing statistically significant structures in PFNS which were deliberately ignored.



^{235}U(n,f) measured at Los Alamos Neutron Science Center (LANSCE) using the Weapons Neutron Research (WNR) facility by Keegan Kelly et al.

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